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OPTICAL CONSTANTS OF BARIUM IN THE VACUUM ULTRAVIOLET REGION

by

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TABLE OF CONTENTS

LIST OF FIGURES

15339

ABSTRACT

An ultra-high vacuum reflectometer has been built to operate at about 5×10^{-10} torr. Barium films have been evaporated and their reflectance measured at two angles, near normal incidence at 17.5° and near grazing incidence at 72.5°. The short wavelength limit was determined by the cut-off of the sapphire windows, namely 1500 Å. With the help of the Fresnel equations, the complex dielectric constant ϵ has been calculated to yield the index of refraction n and the extinction coefficient k. The imaginary part of $(1/\epsilon)$, when plotted against $h\nu$, shows a peak near 7.6 eV and agrees qualitatively with characteristic electron energy loss data. author

CHAPTER I

INTRODUCTION

The measurement of the optical properties of metals in the vacuum ultraviolet is almost completely limited, because of high absorption, lack of polarizers, etc., to reflection techniques. Of these, the most common are the dispersion analysis and the "two angle method." The dispersion analysis requires one experimental measurement of the reflected intensity of arbitrarily polarized, normally incident radiation. This reflectance, which ideally should be measured over an infinite photon energy range, may then be used to relate the real and imaginary parts of the complex index of refraction by means of the Kramers-Kronig dispersion relations.[1] The "two angle method" requires that two reflection measurements be made, with unpolarized light or light of known polarization, at two different angles of incidence. These two values of reflectance then make it possible, in principle, to solve the Fresnel reflection equations for the two unknowns, n, the real part of the index of refraction, and k, the extinction coefficient. The latter technique has been chosen for the present work because the energy range of the available radiation is too narrow to allow a satisfactory dispersion analysis.

The intensity of the reflected beam is given by the Fresnel formulas[2]

$$R_s = \frac{\sin^2(\theta - \chi)}{\sin^2(\theta + \chi)}, \quad R_p = \frac{\tan^2(\theta - \chi)}{\tan^2(\theta + \chi)}, \tag{1}$$

where θ is the angle of incidence, χ the angle of refraction, and the subscripts s and p refer, respectively, to the components polarized perpendicular to and parallel to the plane of incidence. The angles θ and χ are, of course, related by Snell's law

$$\sin\theta = n' \sin\chi , \tag{2}$$

where $n' = n - ik$ is the complex index of refraction. Combination of eqs. (1) and (2) leads to the generalized reflected intensities for unpolarized incident radiation,

$$R_s = \frac{a^2 + b^2 - 2a\cos\theta + \cos^2\theta}{a^2 + b^2 + 2a\cos\theta + \cos^2\theta}, \tag{3}$$

$$R_p = R_s \frac{a^2 + b^2 - 2a\sin\theta\tan\theta + \sin^2\theta\tan^2\theta}{a^2 + b^2 + 2a\sin\theta\tan\theta + \sin^2\theta\tan^2\theta}, \tag{4}$$

$$R = \tfrac{1}{2}(R_s + R_p) = \frac{R_s\,(a^2 + b^2 + \sin^2\theta\tan^2\theta)}{a^2 + b^2 + 2a\sin\theta\tan\theta + \sin^2\theta\tan^2\theta}, \tag{5}$$

where

$$a - ib = n'\cos\chi,$$

$$a^2 + b^2 = [(n^2 - k^2 - \sin^2\theta)^2 + 4n^2k^2]^{\frac{1}{2}},$$

and

$$\sqrt{2}\,a = \{(n^2 - k^2 - \sin^2\theta) + [(n^2 - k^2 - \sin^2\theta)^2 + 4n^2k^2]^{\frac{1}{2}}\}^{\frac{1}{2}}.$$

Unfortunately, the two simultaneous equations which result from the substitution of $R_1(\theta_1)$ and $R_2(\theta_2)$ into eq. 5 cannot be solved to give solutions of the form

$$n = n(R_1, R_2, \theta_1, \theta_2) \text{ and } k = k(R_1, R_2, \theta_1, \theta_2), \tag{6}$$

and a graphical[3,4,5] or tabular technique is required. For this work, the method favored by Ishiguro et al.[5] has been used (see Fig. 1), in which curves of constant n or constant k are plotted in the $(R_{\theta_1}, R_{\theta_2})$ plane. In order to plot these curves, the reflectances $R(\theta_i, n, k)$ are calculated from eq. (5) in steps of $\Delta n = \Delta k = 0.01$ for $0 < n \leq 1$ and $0 < k \leq 1$, and in steps of $\Delta n = \Delta k = 0.1$ for $1 < n, k < 5$. After the experimental reflectance values $R(\theta_i, \omega)$ have been obtained, n and k are given as a function of the angular frequency of the incident radiation, ω, by the pair of curves intersecting at the point $(R(\theta_1, \omega))$. In addition, a program is being written so that the analysis can be made with the aid of a computer.

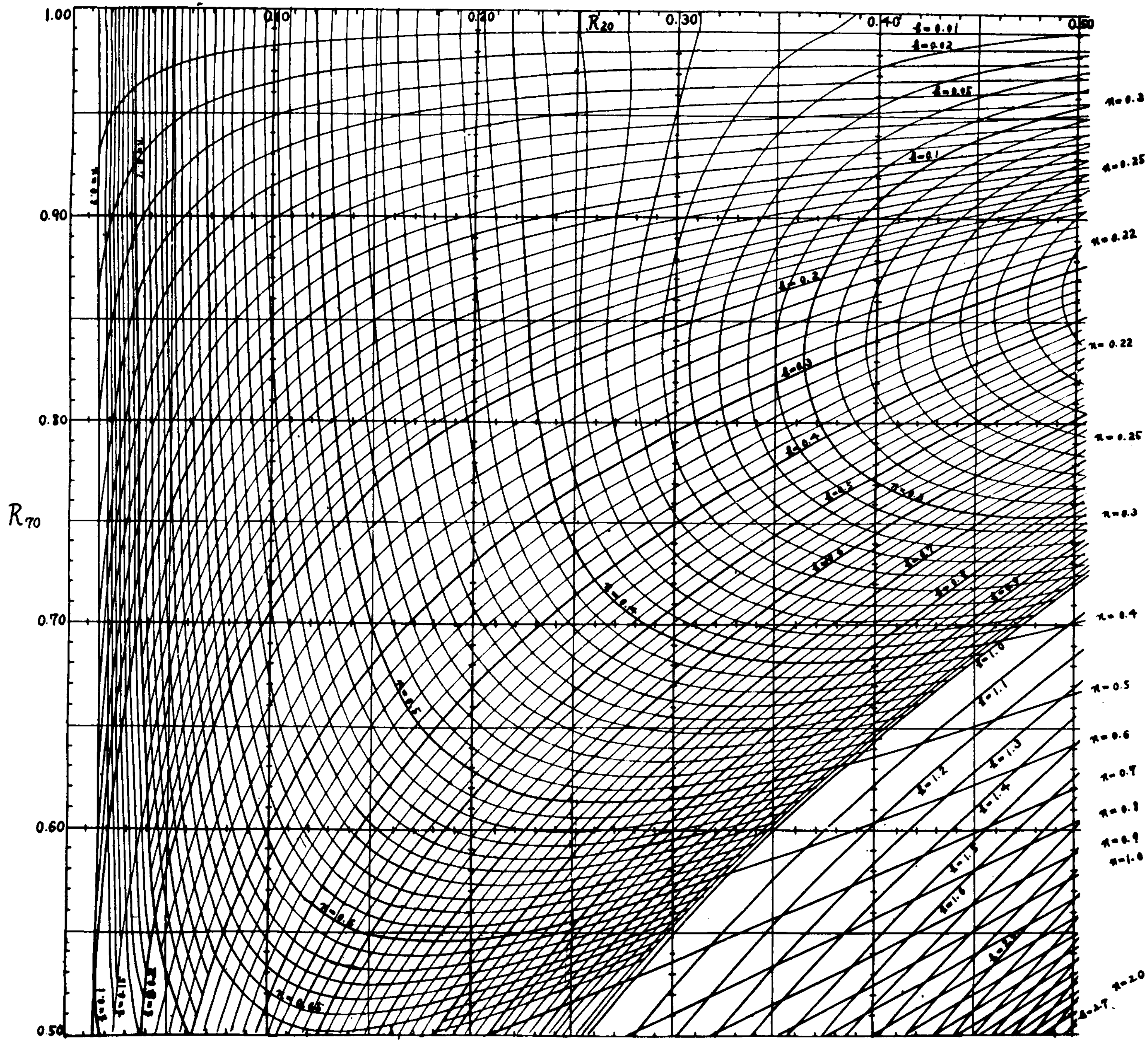

Fig. 1. Ishiguro's Plot (ref. 5) of the Reflectance at 70° versus the Reflectance at 20° for Constant Values of n and k.

CHAPTER II

EXPERIMENTAL ARRANGEMENT AND PROCEDURE

Since reflection measurements are, perforce, extremely sensitive to the surface condition of the sample, an ultra high vacuum reflectometer has been built (Fig. 2), in which reflectivities can be measured at angles of incidence of 17.5° and 72.5°. The ultra high vacuum system is pumped by a 1400 liter/sec oil diffusion pump which is followed by a water cooled baffle and a freon cooled baffle. The combination of these two baffles, which are designed so as to return condensed oil to water cooled parts of the pump, is optically dense and also provides an oil creep (surface migration) barrier. An optically dense, antimigration type liquid nitrogen trap interposes the preliminary baffling and the ultra high vacuum experimental volume. In addition to the oil pumping, titanium sublimators have been incorporated into the ultra high vacuum section. Titanium may thus be sublimated onto the chilled surfaces of the liquid nitrogen trap in order to increase the pumping speed during times of heavy gas load. The 1400 liters/sec diffusion pump is backed by a liquid nitrogen trapped oil sealed rotary pump. Convalex-10 oil[6] is used in both diffusion pumps. The base pressure in the reflectometer is about 5×10^{-10} torr and,

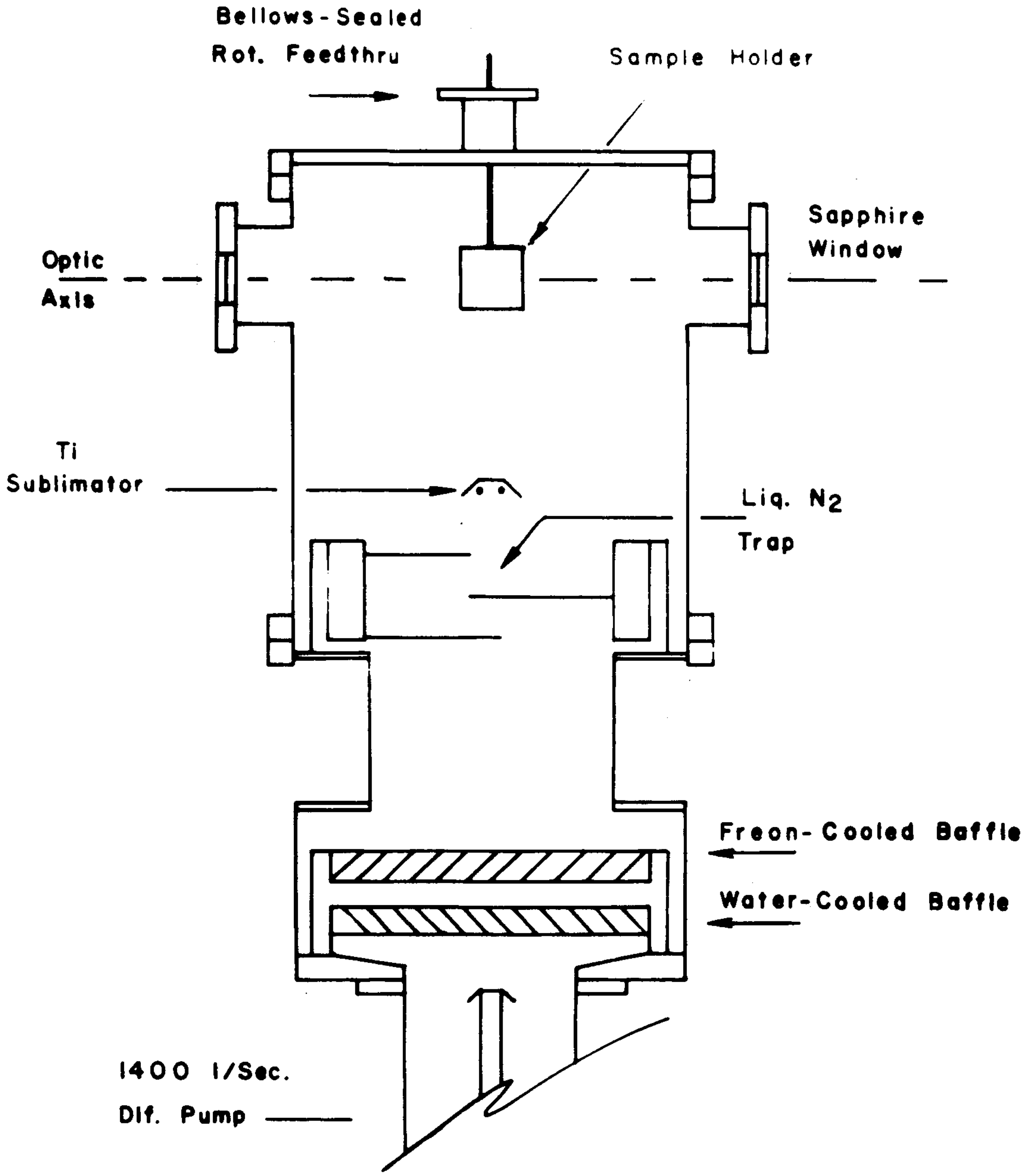

Fig. 2. Ultra-High Vacuum Reflectometer

after initial pumpdown, this pressure can be obtained without repeated bake-outs or use of the Ti sublimators.

In order to accommodate best the requirement of unpolarized incident radiation, a normal incidence vacuum monochromator has been chosen as the source of dispersed ultraviolet radiation. The angle between the entrance and exit arms is 35°. Wavelength scans are made by rotating the grating about a vertical axis through its center. When used in the first outside order with a 1440 grooves/mm grating, the angle of incidence at the grating of the undispersed radiation varies between 11.0° and 4.4° in the wavelength range between 1500 and 3000Å. The problem of polarization by reflection is thus minimized by the small angle of incidence. Although the focusing properties of such an instrument are not as good as those of the Seya[7] type monochromator, it is possible to optimize the geometry so that, for the above mentioned range and with a grating having a 1 meter radius of curvature, the position of the focused image is always within 3 mm of the exit slit. This is done by setting the entrance slit at 78.740 cm from the grating and the exit slit 124.59 cm from the grating. The distance from the grating to the focused image varies between 124.30 and 124.8 cm for this wavelength range. For a Seya monochromator, the corresponding defocusing would be about 0.1 mm.

In Fig. 3, a schematic representation of the experimental arrangement, the monochromator is shown in conjunction

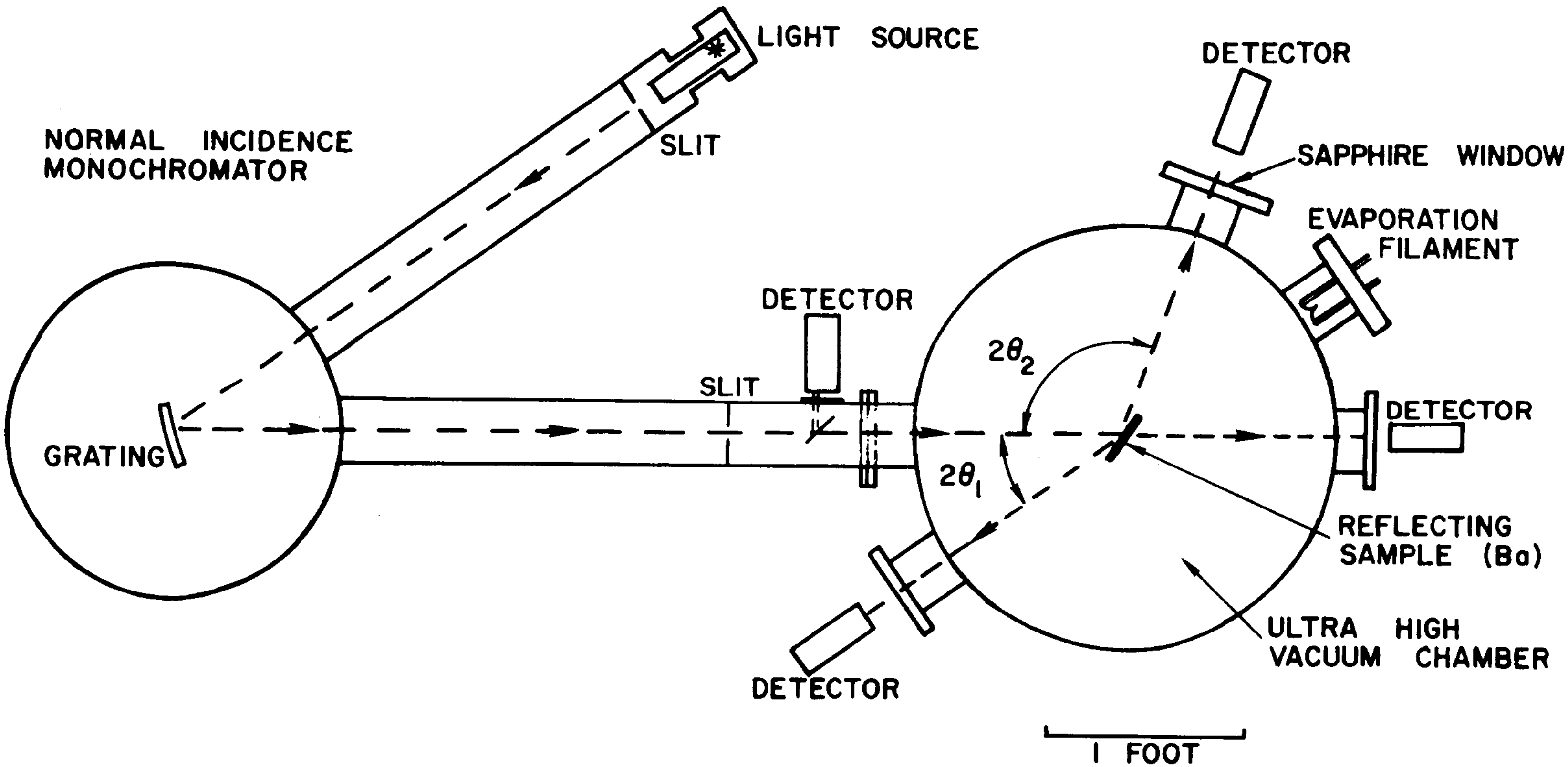

Fig. 3. Vacuum Monochromator Coupled to Ultra-High Vacuum Reflectometer. Radiation from the light source passes through the entrance slit and illuminates the grating which disperses it. Consequently, monochromatic light emerges from the exit slit and passes through a sapphire window into the ultra-high vacuum chamber, where the sample surface (Ba) reflects at near normal incidence ($\theta_1 = 17.5^\circ$) to a photomultiplier detector or at near grazing incidence ($\theta_2 = 72.5^\circ$) with the detector moved to the new position. The incident intensity is measured by moving the sample up and out of the optic axis. The sample is deposited by evaporation.

with the reflectometer. It is sealed to the reflectometer by means of an elastomer gasket at the flange near the exit slit. The presence of a sapphire window in this flange isolates the ultra high vacuum of the reflectometer from the relatively poor vacuum of the monochromator, yet permits the transmission of wavelengths longer than 1500Å. The light source is a continuously pumped dc glow discharge in hydrogen, which provides a line spectrum below 1700Å and a continuum at longer wavelengths. The detector is a sodium salicylate coated photomultiplier in a mounting which can be interchanged between the indicated flanges so as to measure the incident intensity(with the sample removed in an upward direction from the optic axis) and the intensities reflected at the angles $\theta_1 = 17.5^\circ$ and $\theta_2 = 72.5^\circ$. The photomultiplier housing is evacuated by means of a mechanical pump in order to permit transmission of wavelengths shorter than 2000Å.

The barium is evaporated from 0.005 inch wall thickness tantalum boats. With a boat of this thickness an evaporation current of about 100 amp is necessary. The evaporation takes place slowly, and a time of about 15 minutes is required to obtain a visually detectable film. The substrate, which is maintained at room temperature, consists of a polished stainless steel plate (or, alternatively, a microscope glass slide) attached to a bellows-sealed rotary feedthrough.

Prior to any reflection measurements the barium evaporation boats and titanium sublimation filaments are outgassed, and the system is allowed to return to its base pressure. A fresh layer of titanium is then sublimated onto the cold trap and the barium evaporation is performed. The maximum pressure during the evaporation is in the 10^{-6} torr range and returns to the 10^{-9} torr range within five minutes. Reflectivity measurements ($1500Å < \lambda < 3000Å$) at the two angles, 17.5° and 72.5°, are then taken. Repeated measurements are made on a single film to investigate changes which occur due to aging of the film.

CHAPTER III

RESULTS AND DISCUSSION

Preliminary results, taken in Dec. 1964, are shown in Fig. 4, where reflectance measurements at 17.5° and at 72.5° are plotted versus $h\nu$. Fig. 5 gives the corresponding n and k values, and Fig. 6 those for $\mathrm{Im}(1/\epsilon)$. Figs. 7, 8 and 9 show corresponding results of R, n and k, and $\mathrm{Im}(1/\epsilon)$, all versus $h\nu$; these data, taken in July 1965, represent probably the most reliable optical measurements to date. The quantity $\mathrm{Im}(1/\epsilon)$ is equal to $2nk/(n^2 + k^2)^2$, where ϵ is the complex dielectric constant; it is of interest because it is proportional to the cross section for collective electron excitation.[8] Robins and Best[9] have reported an electron characteristic loss of 6.5 eV in barium, shown by a downward arrow marked e^- in Figs. 6 and 9. Thus if the optical constants are determined primarily by free electron contributions, the imaginary part of the reciprocal of the dielectric constant should show a maximum at this energy. Figs. 6 and 9 show a peak at 6.25 eV and at 7.6 eV, respectively, which is in qualitative agreement with this assumption. The later, more reliable data have indicated, however, that the $\mathrm{Im}(1/\epsilon)$ - curve is narrower than the same curves determined earlier.

As the film ages in the ultra high vacuum system, the maximum of $\mathrm{Im}(1/\epsilon)$ becomes broader and lower until it dis-

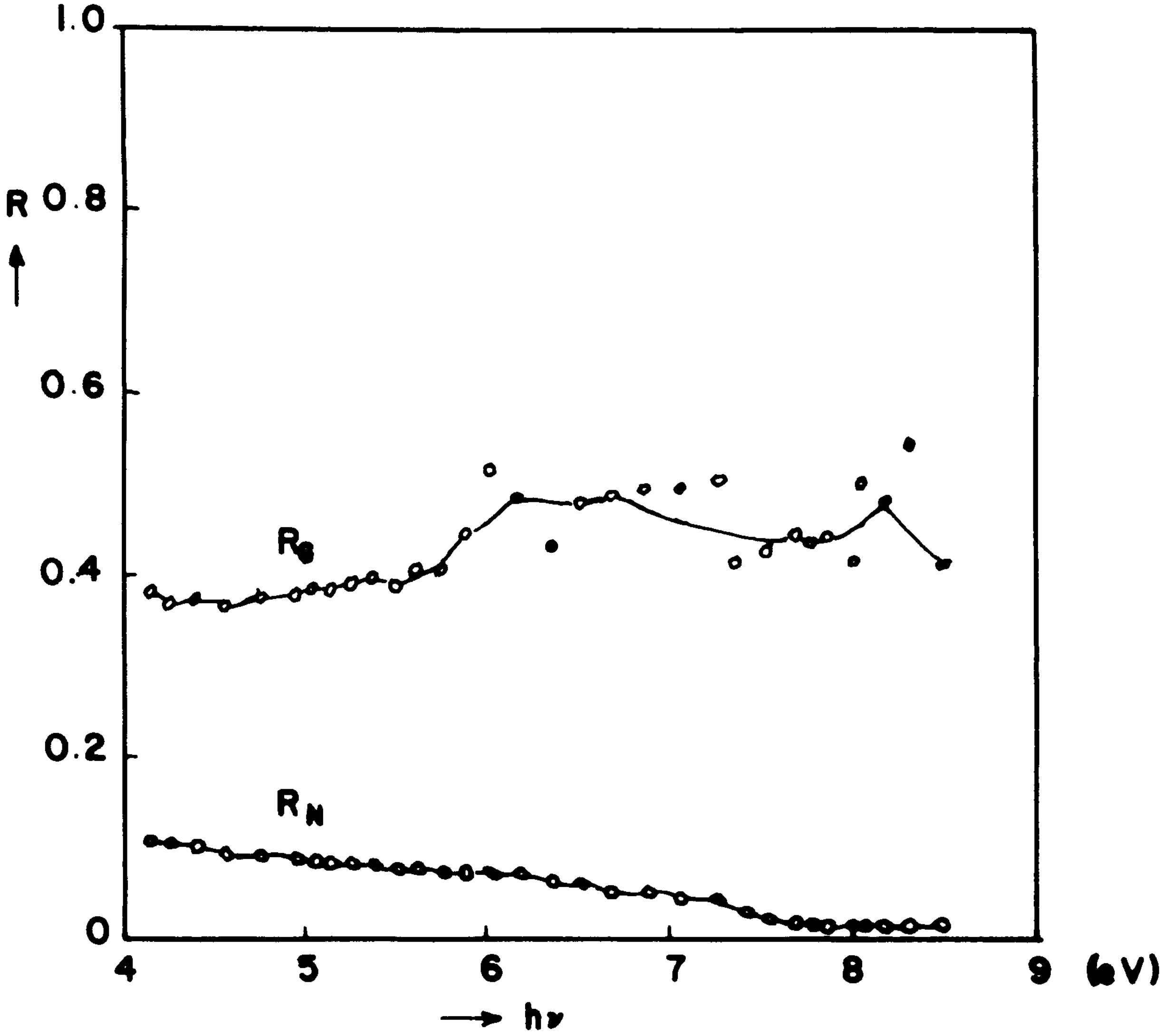

Fig. 4. Dec. 1964: Barium Reflectance Measurements taken at two Angles, R_N at 17.5° and R_G at 72.5°.

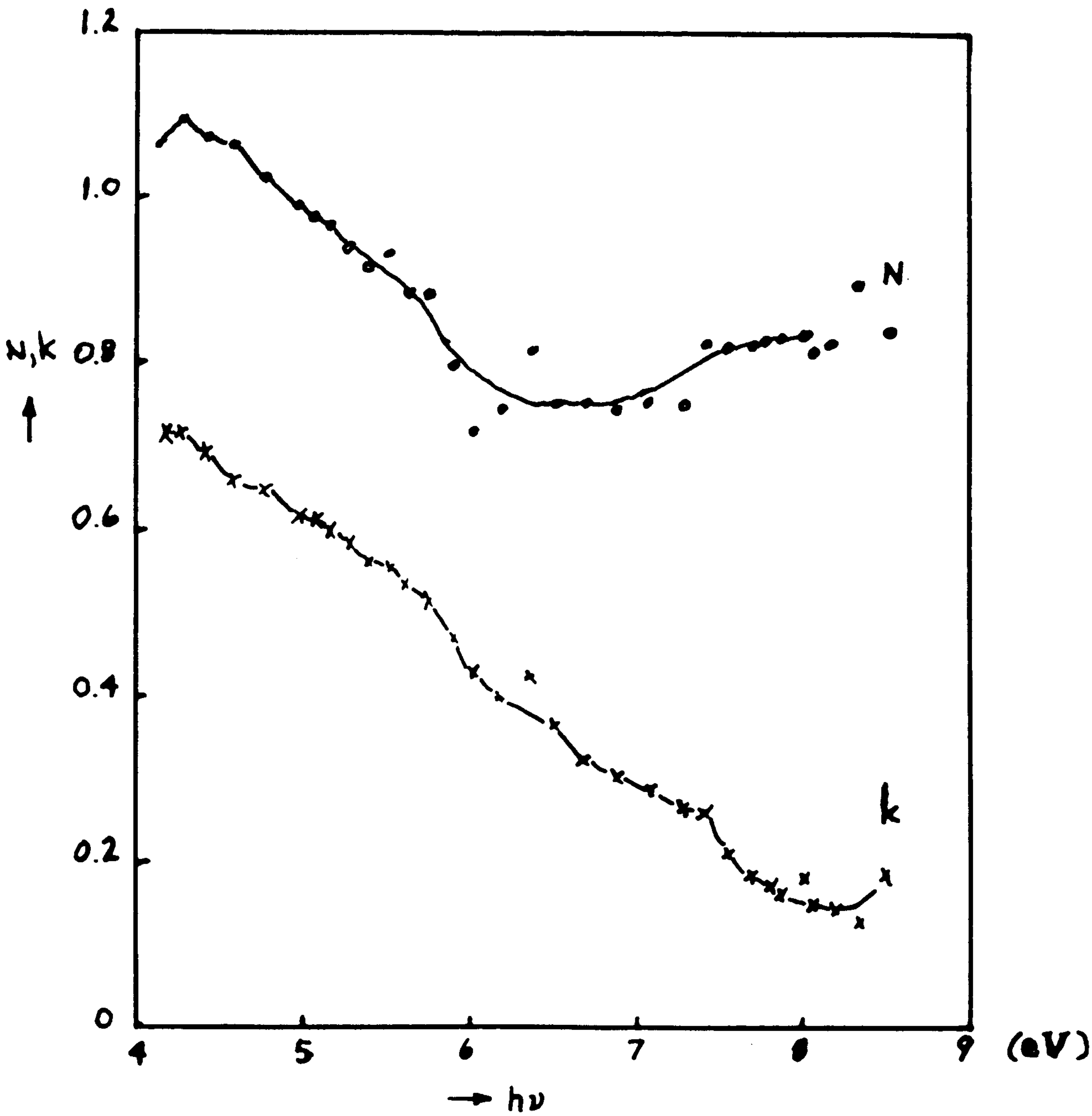

Fig. 5. Dec. 1964: n and k Values versus hν for Barium Calculated from Reflectance Data in Fig. 4.

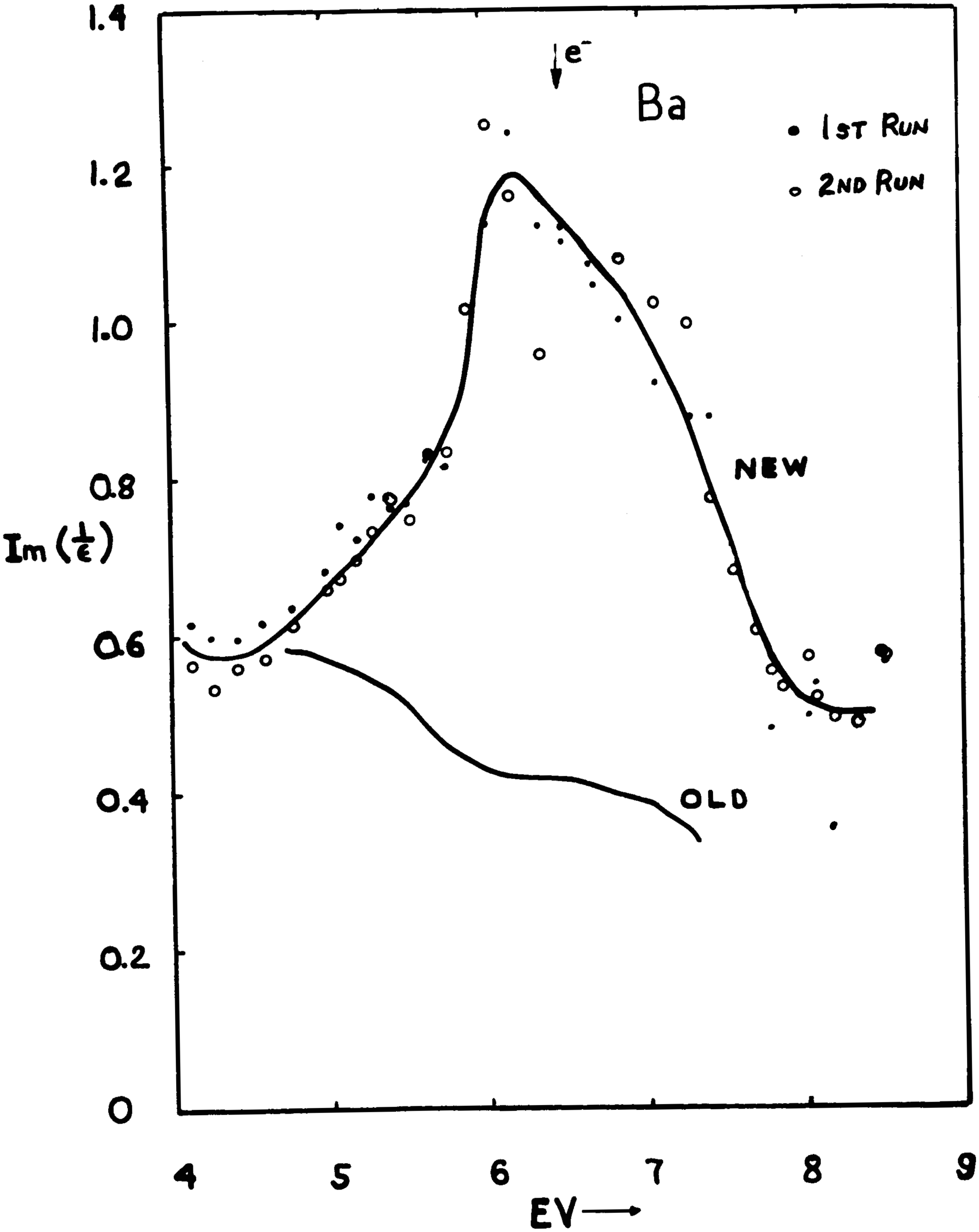

Fig. 6. Dec. 1964: Values of Im (1/ε) versus hν for Barium Calculated from Reflectance Data in Fig. 4.

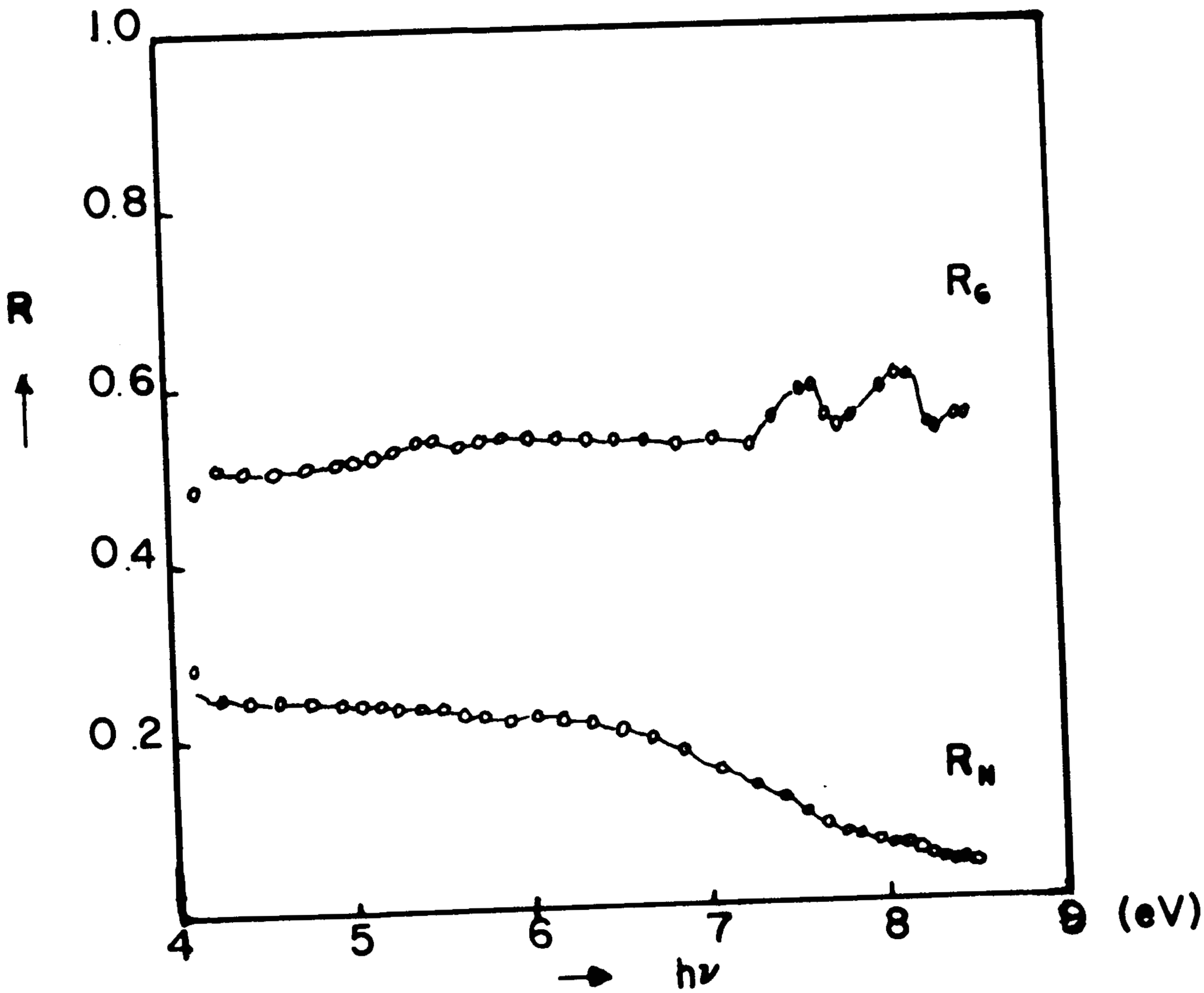

Fig. 7. July 1965: Barium Reflectance Measurements taken at two Angles, R_N at 17.5° and R_G at 72.5°

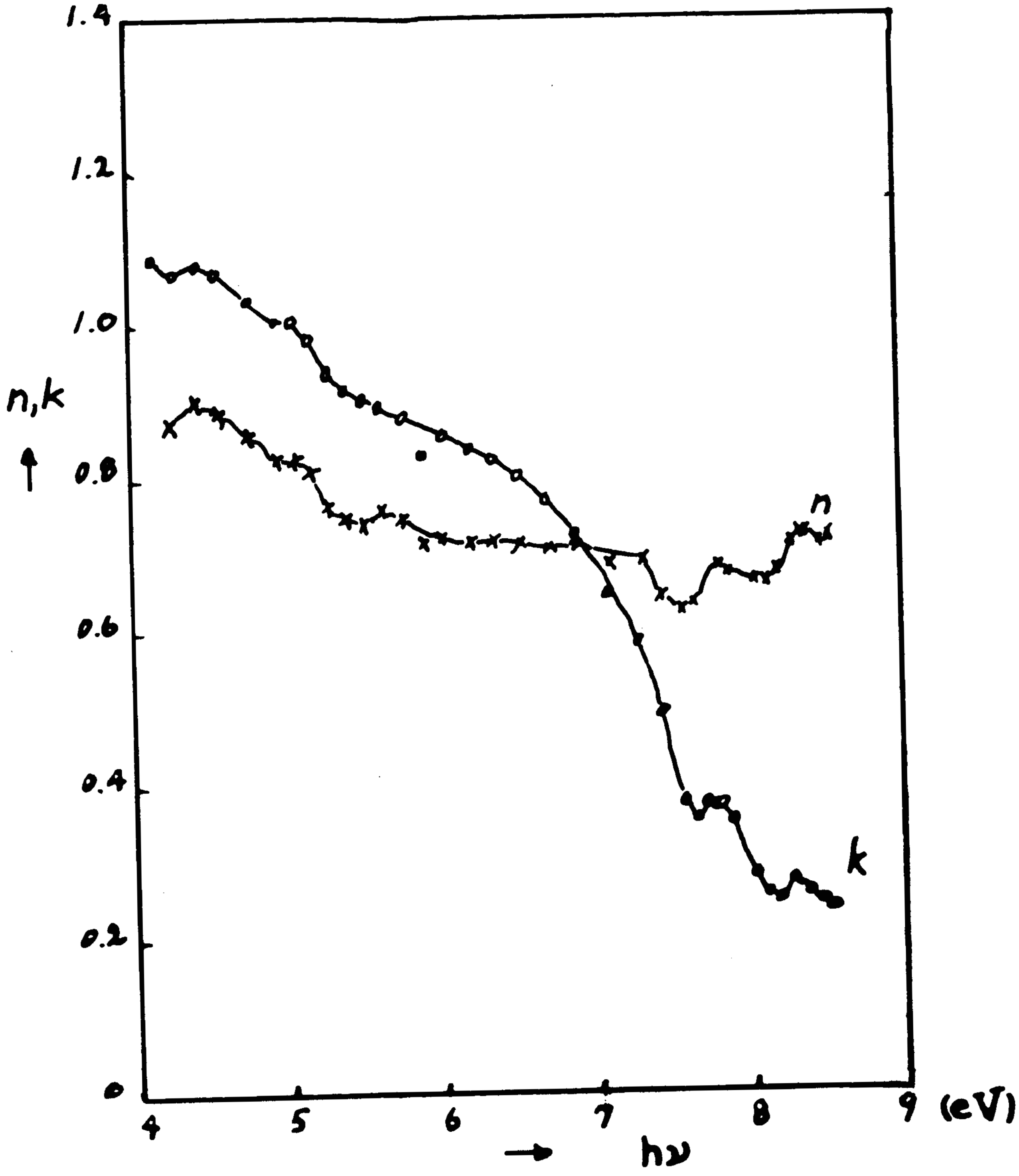

Fig. 8. July 1965: n and k Values versus hν for Barium Calculated from the Reflectance Data in Fig. 7.

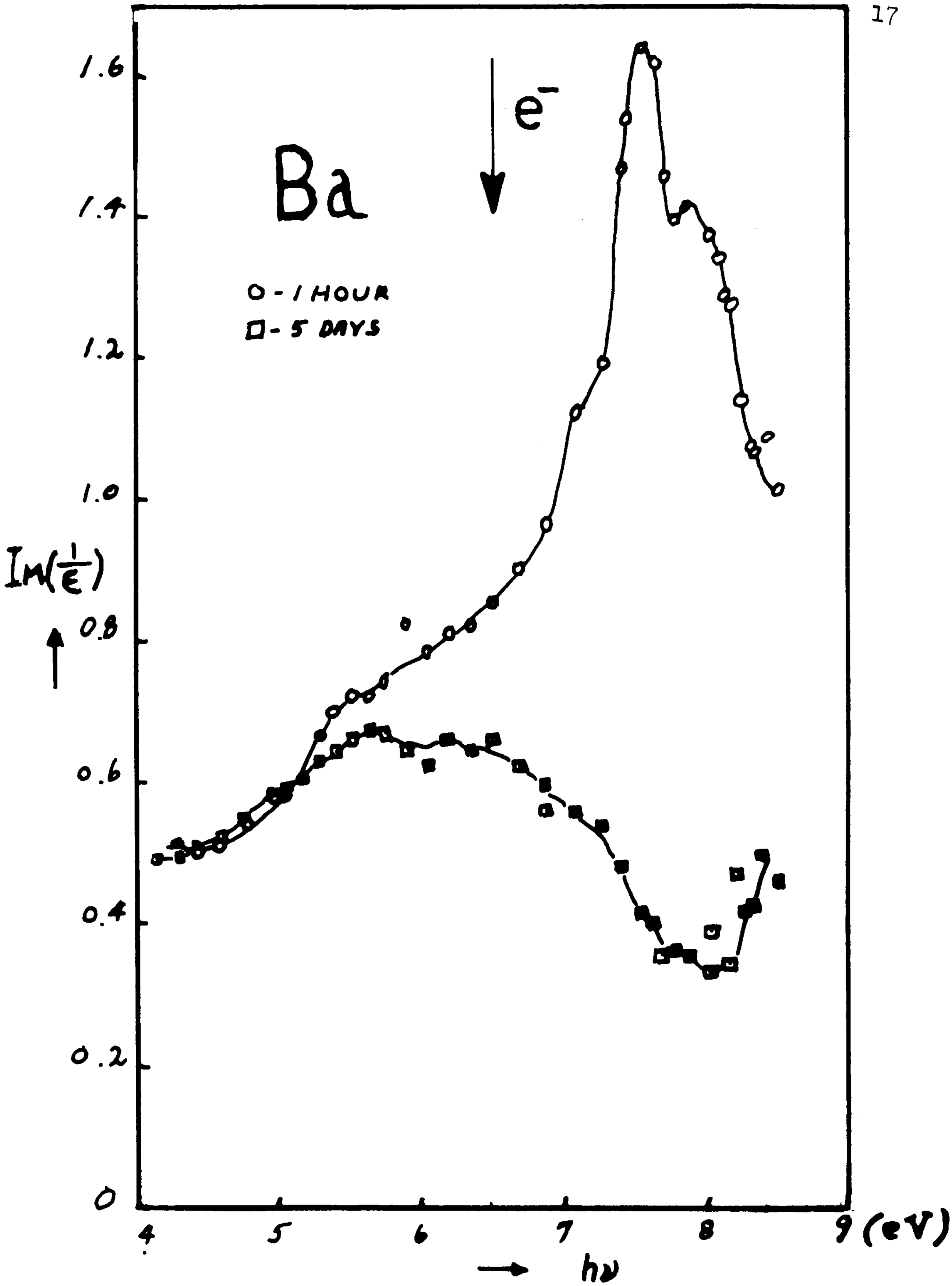

Fig. 9. July 1965: Values of Im $(1/\varepsilon)$ versus $h\nu$ for Barium Calculated from the Reflectance Data in Fig. 7.

appears altogether. The lower curves in Figs. 6 and 9 represent this quantity measured after the barium film had been exposed to a pressure of about 1 x 10^{-9} for eight days.

Between Dec. 1964 and July 1965, a total of 20 newly evaporated barium surfaces have been investigated in 45 separate runs of reflectance determinations versus wavelength. In nearly all cases, the $\mathrm{Im}(1/\epsilon)$ versus $h\nu$ curves show a maximum located between that of the earlier results of Dec. 1964 (Fig. 6) and that of the last data of July 1965 (Fig. 9). In addition, measurement techniques have sufficiently improved to suggest details about the $\mathrm{Im}(1/\epsilon)$ curve, such as possible secondary peaks of as yet unknown origin.

Even though it is well known, that H_2 and CO are the principal residual gases in an ultra-high vacuum system as used in this research, it is not clear at this time as to what contaminations contribute to the optical deterioration of these barium surfaces. More work in this direction is necessary.

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